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Self-organization and properties of dispersed systems based on dilute aqueous solutions of (*S*)- and (*R*)-lysine

Irina S. Ryzhkina,*[a] Svetlana Yu. Sergeeva,[a] Yuliya V. Kiseleva,[a] Alina P. Timosheva,[a] Olga A. Salakhutdinova,[a] Maxim D. Shevelev[a,b] and Alexander I. Konovalov[a]

[a] *A. E. Arbuzov Institute of Organic and Physical Chemistry, Kazan Scientific Center, Russian Academy of Sciences, 420088 Kazan, Russian Federation. Fax: +7 843 273 1872; e-mail: ryzhkina@iopc.ru*

[b] *Institute of Chemistry, Kazan Federal University, 420008 Kazan, Russian Federation*

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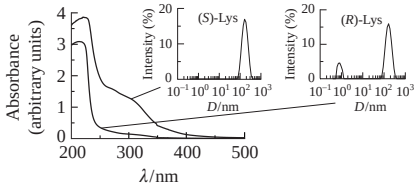
In a concentration range of 1.0–1.0 × 10^{-17} mol dm^{-3}, aqueous systems based on (*S*)- and (*R*)-lysine vary in the ability to form a dispersed phase (domains and nanoassociates) hundreds nanometers in size with different UV absorption spectra of these systems.



The nonmonotonic concentration dependence of the effects of highly diluted aqueous solutions (1.0 × 10^{-20}–1.0 × 10^{-6} mol dm^{-3}) of biologically active compounds (BACs) attracts considerable attention. However, the application of this phenomenon in pharmacology and medicine is limited by the lack of an explanation of its physicochemical nature.[1]

Recently, based on experimental data,[2,3] it was hypothesized that the nonmonotonic response of biosystems to the effect of highly diluted BAC solutions, the effect of silent zones and the bioeffect sign reversal (hormesis) can be explained by the fact that so-called highly diluted solutions are nanoheterogeneous dispersed systems that undergo a supramolecular domain[4,5]–nanoassociate[2,3] rearrangement in the disperse phase on dilution, which is accompanied by changes in the physicochemical and biological properties of the system.

The nanoassociates[2,3,6,7] are fractal objects hundreds of nanometers in dimension that are formed in dilute aqueous and aqueous-organic systems and involve solute and solvent molecules. They have an electrically charged interface and enhanced viscosity in comparison with the dispersing medium. Domains are generally formed at calculated concentrations of 1–10^{-5} mol dm^{-3}, while nanoassociates are formed at much lower concentrations of 10^{-6}–10^{-20} mol dm^{-3}.

One of the main differences between domains and nanoassociates is that domains are formed both in the presence and in the absence of low-frequency background electromagnetic fields (hypoelectromagnetic conditions).[2,3] Water and aqueous solutions are very sensitive to the impact of weak physical and chemical factors.[8,9] Most likely, it is owing to the fundamental phenomenon that nanoassociates have the unique capability to be formed in the presence of low-frequency electromagnetic fields even at ultra-low BAC concentrations.

Currently, experimental[4,10] and theoretical[8,11] studies of the domains and nanoassociates and their role in the operation of complex biological systems are in progress. The effect of the chemical and spatial BAC structure on the self-organization of aqueous systems is of importance. Therefore, we studied the self-organization and properties of aqueous systems based on α-amino acids[12] and structures containing their fragments.[13] All α-amino acids found in living organisms, except for glycine, are optically active and have enantiomeric L- and D-forms or, in terms of absolute configuration, (*S*)- and (*R*)-forms.[14] The enantiomers not only have different spatial structures of molecules but also show pronounced distinctions in their biological properties due to differences in permeation through biomembranes, in reactions with enzymes, *etc.*[14,15]

In this work, we used dilute (1.0–1.0 × 10^{-18} mol dm^{-3}) aqueous solutions of (*S*)- and (*R*)-lysine (Lys) (**1** and **2**), an essential proteogenic amino acid with a side chain protonated in a neutral pH range,[14] as test materials. (*S*)-Lys has a broad spectrum of biological activity (including the synthesis of antibodies, hormones and enzymes, collagen formation and restoration of tissues) and affects the nervous and immune regulatory systems. The low-concentration solutions of (*S*)-Lys alone and in pharmaceutical formulations[15] also exhibit biological properties and therapeutic effects. (*R*)-Lys lacks these properties.

The preparation of (*S*)- and (*R*)-Lys solutions and studies of the self-organization and properties of the solutions by physicochemical methods were reported previously[2,3,7] (see Online Supplementary Materials).

A dynamic light scattering (DLS) analysis of the aqueous solutions of **1** and **2** (Figure 1) has revealed that the solutions of both enantiomers are nanoheterogeneous systems similarly to BACs studied previously.[2,3,7] However, the self-organization processes in these solutions in the test concentration range have certain differences.

Judging from the light scattering intensity (*I*) at 1.0 mol dm^{-3} [Figure 1(*a*),(*b*)], domains with sizes (*D*) of about 250 nm predominated in the system based on (*S*)-Lys (93%), whereas particles with diameters around 1 nm predominated in the (*R*)-Lys based system (60%). These particles (micelle-like structures[5]) are dynamic fractal objects mainly consisting of solute (~60%) and water molecules.

On dilution of the (*S*)-Lys based system to 2.5 × 10^{-1}, 1.0 × 10^{-1} mol dm^{-3} and below, the intensity size distribution becomes nearly unimodal, and a unimodal distribution was observed in a range from 1.0 × 10^{-2} to 1.0 × 10^{-7} mol dm^{-3} (see Online Supplementary Materials). The process of self-organization